

REMARKS

The claims in the case are claims 1-17 and 19-29. The claims have been amended to eliminate multiple dependency and to put them in better form for U.S. filing.

No new matter is included.

Favorable action is solicited.

Respectfully submitted,

KEIL & WEINKAUF



Herbert B. Keil

Reg. No. 18,967

1350 Connecticut Ave., N.W.
Washington, D.C. 20036

(202)659-0100

CLAIMS 53622

1. (currently amended) A process for preparing optionally alkyl-substituted 1,4-butanediol by two-stage catalytic hydrogenation in the gas phase of C₄-dicarboxylic acids and/or of derivatives thereof having the following steps:
 - a) introducing a gas stream of a C₄-dicarboxylic acid or of a derivative thereof at from 200 to 300°C and from 10 to 100 bar into a first reactor or into a first reaction zone of a reactor and catalytically hydrogenating it in the gas phase to a product which contains mainly optionally alkyl-substituted γ -butyrolactone;
 - b) introducing the product stream obtained in this way into a second reactor or into a second reaction zone of a reactor at a temperature of from 140°C to 260°C and catalytically hydrogenating it in the gas phase to optionally alkyl-substituted 1,4-butanediol;steps a) and b) being carried out at the same pressure;
- c) removing the desired product from intermediates, by-products and any unconverted reactant;
- d) optionally recycling unconverted intermediates into one or both hydrogenation states,

said hydrogenation stages each using a catalyst which comprises $\leq 95\%$ by weight of CuO, and $\geq 5\%$, preferably from 5 to 95% by weight, in particular from 10 to 80% by weight, of CuO, and $\geq 5\%$ by weight, preferably from 5 to 95% by weight, in particular from 20 to 90% by weight, of an oxidic support, and the product mixture removed from the first hydrogenation

CLAIMS 53622

stage being introduced without further purification into the second hydrogenation stage.

2. (currently amended) The A process as claimed in claim 1, wherein the entrance temperature into the second reactor or into the second reaction zone of a reactor is lower than the entrance temperature into the first reactor or into the first reaction zone.
3. (currently amended) The A process as claimed in claim 1 or 2, wherein the entrance temperature into the first reactor is from 235 to 270°C and the entrance temperature into the second reactor is from 160°C to 225°C, in particular from 180 to 200°C.
4. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 3~~, wherein the hot spot temperature in the first reactor is from 210 to 310°C, ~~preferably from 245 to 280°C~~, and the process is carried out in such a manner that the hot spot temperature is above the entrance temperature and the exit temperature of the reaction gases, and is from 5 to 30°C, ~~in particular from 5 to 15°C, more preferably from 5 to 10°C~~, above the entrance temperature.
5. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 4~~, wherein the pressure in both the first hydrogenation stage and the second hydrogenation stage is from 10 to 100 bar, ~~preferably from 15 to 50 bar~~.
6. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 5~~, wherein the catalyst hourly space velocity of the first hydrogenation stage is in the range from 0.02 to 1, ~~in particular from 0.05 to 0.5~~, kg of reactant/l catalyst •

CLAIMS 53622

hour, and the catalyst hourly space velocity of the second hydrogenation stage is in the range from 0.02 to 1.5, in particular from 0.1 to 1, kg of reactant/l of catalyst • hour.

7. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 6~~, wherein the hydrogen/reactant molar ratio in both reaction stages is ≥ 5 [[,]] preferably from 20 to 600.
8. (currently amended) The A process as claimed in claim 7, wherein the hydrogen/reactant ratio in the first stage hydrogenation is from 20 to 200[[,]] preferably from 40 to 150, in particular from 50 to 100.
9. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 8~~, wherein the reactors used are selected from the group consisting of tubular reactors, shaft reactors, reactors having internal heat removal means, preferably tube bundle reactors and fluidized bed reactor.
10. (currently amended) The A process as claimed in claim 9, wherein a tube bundle reactor is used in the first hydrogenation stage.
11. (currently amended) The A process as claimed in claim 9 ~~or 10~~, wherein a shaft reactor is used in the second hydrogenation stage.
12. (currently amended) The A process as claimed in claim 1 ~~any of claim 1 to 11~~, wherein more than one reactor connected in parallel or in series is used in the first and/or second hydrogenation stage.
13. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 12~~, wherein a two-zone reactor is used.

CLAIMS 53622

14. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 13~~, wherein the process is carried out in an apparatus having a hydrogenation cycle and a compressor.
15. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 14~~, wherein the support material of the catalyst is selected from the group of ZnO, Al₂O₃, SiO₂, TiO₂, ZrO₂, CeO₂, MgO, CaO, SrO, BaO and Mn₂O₃ and mixtures thereof, preferably from the group of ZnO/Al₂O₃ mixtures, the delta-, theta-, alpha- and eta-modifications of Al₂O₃ and also mixtures which comprise at least one component each firstly from the group of SiO₂, TiO₂, ZrO₂, and secondly from the group of ZnO, MgO, CaO, SrO and BaO.
16. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 15~~, wherein the support material is selected from ZnO, ZnO/Al₂O₃ mixtures in a weight ratio of from 100:1 to 1:2 and mixtures SiO₂ with MgO, CaO and/or ZnO in a weight ratio of 200:1 to 1:1.
17. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 16~~, wherein the catalyst comprises one or more further metals, ~~preferably Pd, or a compound of one or more further metals, preferably an oxide,~~ from groups 1 to 14 of the Periodic Table.
18. (canceled)
19. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 18~~, wherein BET surface area of the copper catalysts in the oxidic state is from 10 to 300 m²/g, ~~preferably from 15 to 175 m²/g, in particular from 20 to 150 m²/g.~~

CLAIMS 53622

20. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 19~~, wherein the copper surface area of the reduced catalyst in the installed state is > 0.2 m²/g, ~~preferably > 1 m²/g, in particular > 2 m²/g.~~
21. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 20~~, wherein the catalysts used in the first and second reactors are identical or different, preferably different.
22. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 24~~, wherein the shaped bodies of the catalyst used have a pore volume of ≥ 0.01 ml/g for pore diameter of > 50 nm, ~~preferably of ≥ 0.025 ml/g for pore diameter of > 100 nm and in particular of ≥ 0.05 ml/g for pore diameters of > 200 nm.~~
23. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 12~~, wherein the shaped bodies of the catalyst used have a ratio of micropores having a diameter of > 50 nm to the total pore volume for pores having a diameter of > 4 nm of > 10%, ~~preferably > 20%, in particular > 30%.~~
24. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 23~~, wherein the reactant used in the reaction is maleic anhydride.
25. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 24~~, wherein maleic anhydride is used which has been prepared by oxidizing benzene, C₄-olefins or n-butane, and the crude maleic anhydride obtained by oxidation has been extracted from the crude product mixture using a solvent (absorbent) and then stripped from this solvent using hydrogen.
26. (currently amended) A process as claimed in claim 1 ~~any of claims 1 to 25~~,

CLAIMS 53622

wherein the absorbent is selected from the group consisting of tricresyl phosphate, dibutyl maleate, high molecular weight waxes, aromatic hydrocarbons having a molecular weight of from 150 to 400 and boiling point above 140°C, ~~preferably dibenzene, di-C₁-C₄-alkyl esters of aromatic and aliphatic dicarboxylic acids, preferably dimethyl 2,3-naphthalenedicarboxylate and/or dimethyl 1,4-cyclohexanedicarboxylate,~~ methyl esters of long-chain fatty acids having from 14 to 30 carbon atoms, high-boiling ethers, ~~preferably dimethyl ethers of polyethylene glycol, preferably of tetraethylene glycol,~~ and alkyl phthalates and dialkyl phthalates having C₁-C₁₈-alkyl groups, ~~preferably from the group of dimethyl phthalate, diethyl phthalate, dibutyl phthalate, di-n-propyl and diisopropyl phthalate, undecyl phthalate, diundecyl phthalate, methyl phthalate, ethyl phthalate, butyl phthalate, n-propyl and isopropyl phthalate.~~

27. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 26,~~ wherein the maleic anhydride is stripped from the absorbent under reduced pressure of pressures which correspond to the hydrogenation pressure or are a maximum of 10% above this pressure.
28. (currently amended) The A process as claimed in claim 1 ~~any of claims 1 to 27,~~ which is carried out batchwise, semicontinuously or continuously, preferably continuously.
29. (new) The process as claimed in claim 1, wherein the catalyst is used in the form of shaped bodies, preferably in the form of extrudates, ribbed extrudates, tablets, rings, spheres or spall.